LA-UR-02-4909

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Title: Singlet and triplet states of trions in ZincSelenide-based quantum wells probed by magnetic fields to 50 tesla

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Submitted to: International Conference of the Physics of Semiconductors
Edinburgh, Scotland
July 29-Aug 3, 2002

Singlet and triplet states of trions in ZnSe-based quantum wells probed by magnetic fields to 50 Tesla

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Abstract. Singlet and triplet states of positively (X^+) and negatively (X^-) charged excitons in ZnSe-based quantum wells have been studied by means of photoluminescence in pulsed magnetic fields up to 50 T. The binding energy of the X^- singlet state shows a monotonic increase with magnetic field with a tendency to saturation, while that of the X^+ slightly decreases. The triplet X^+ and X^- states, being unbound at zero magnetic field, noticeably increase their binding energy in high magnetic fields. The experimental evidence for the interaction between the triplet and singlet states of trions leading to their anticrossing in magnetic fields has been found.

1. Introduction

Charged excitons (or trions) are exciton complexes consisting of three particles. Two electrons and one hole form a negatively charged exciton X^-. Two holes and one electron can be organized in a positively charged exciton X^+. Trion complexes in bulk semiconductors, i.e. in three dimensions, are fragile, but become stable in low-dimensional systems (for instance, in quantum wells).

II-VI semiconductor heterostructures and especially ZnSe-based quantum wells (QW's) are attractive for the investigation of charged excitons because of the strong Coulomb interaction. The exciton binding energy (with which trion binding energy is scaled) of 20 meV in bulk ZnSe considerably exceeds the 4.2 meV exciton Rydberg in bulk GaAs. The small size of the exciton state in ZnSe structures requires, however, even very high magnetic fields to resolve the energy and spin structure of trion states. We report here magneto-optical studies performed in pulsed magnetic fields to 50 T.

2. Experimental

ZnSe-based quantum well heterostructures with a binary material of QW were grown by molecular-beam epitaxy on (100)-oriented GaAs substrates. Studied structures contain single quantum wells, with thickness varying from 29 to 190 Å. Two different barrier materials were used, namely $Zn_{0.89}Mg_{0.11}S_{0.18}Se_{0.82}$, and $Zn_{0.82}Be_{0.08}Mg_{0.10}Se$. Parameters of the barrier materials were chosen with an aim to make them lattice-matched to GaAs substrates, which allow growing QW's of a very high structural quality. The detailed analysis of optical properties of these structures is published in Ref. [Ast02a].

The structures with the two-dimensional electron gas (2DEG) used in this study were nominally undoped. A background carrier density in them was tuned by an additional above-barrier illumination [Ast02a]. The range of tuning depends on the QW width, allowing in the widest QW to vary electron density from 5×10^{9} to 10^{11} cm^{-2}. The sample, containing the two-dimensional hole gas (2DHG), was *p*-type doped with nitrogen (RF plasma cell at a power of 350 W and a background pressure of 5×10^{-6} Torr). In this sample, symmetric doping was achieved by uniform doping of barriers excluding 30-Å-thick spacer layers. The density of the 2DHG in the QW of this sample is about $n_h \approx 3\times10^{10}$ cm^{-2} and was insensitive to additional illumination.

Photoluminescence (PL) technique was exploited for experimental study of trion properties in high magnetic field applied along the growth direction (Faraday geometry). Experiments were provided in a capacitor-driven 50 T mid-pulse magnet (~400 ms decay) at the National High Magnetic Field Laboratory (Los Alamos, USA). A complete set of field-dependent PL spectra was collected at a low temperature (T=1.6 K) during each magnetic field pulse (for details see Ref. [Croo99]). PL was excited by He-Cd laser of low power (about 1 mW). For optical access to the sample fiber optics were used, and emitted light was detected in two circular polarizations allowing resolving spin components of excitons and trions.

3. Photoluminescence of trions in high magnetic fields

Figure 1 displays typical PL spectra of $ZnSe/Zn_{0.82}Be_{0.08}Mg_{0.10}Se$ QW containing diluted 2DEG. In zero magnetic field (lower spectrum) two lines are observed. The high-energy line is due to exciton (X) recombination in the QW. And the low-energy line is attributed to a negatively charged exciton (X_s^-), which is constructed of an electron bound to neutral exciton [Cox93]. Identification of charged exciton lines in optical spectra of ZnSe-based QW's was done in Ref. [Ast99]. An energy distance between these lines of 5.3 meV corresponds to the binding energy of negatively charged exciton (or trion). This value is only 18% of exciton binding energy, which is 30 meV (a detailed analysis of trion binding energy versus QW width, electron concentration and magnetic field can be found in Ref. [Ast02a]).

Two different spin configurations of the two electrons (S^e) involved in the negatively charged exciton are possible. They form the singlet (X_s^-) and the triplet

(X_t^-) states of trions. The former corresponds to the total spin of the two electrons $S^e=0$ with a projection $S_z^e=0$. This singlet state, with the antiparallel orientation of electron spins, is the ground state of trions at zero magnetic field. The triplet state X_t^- is formed with electrons having $S^e=1$ and, therefore, with three possible projections $S_z^e=0, \pm 1$. The triplet state of negatively charged exciton is unbound in the absence of magnetic field. External magnetic field makes the X_t^- stable, and in high magnetic fields (above few tens of Tesla) the triplet state of trion crosses the singlet state and becomes the lowest energy state. [Whi97]).

The behaviour of X_t^- in magnetic fields is seen in Fig. 1 (upper curves). At $B=20$ T a new line is observed between the exciton and the singlet state X_s^- of the trion. This line is ascribed to the triplet state of the negatively charged exciton, X_t^-. An identification of this line is based on its energy shift in magnetic fields (see Figs. 3 and 4), which is similar to that in Refs. [Shi95a, Fin96].

In Fig. 2 the PL of $ZnSe/Zn_{0.89}Mg_{0.11}S_{0.18}Se_{0.82}$ QW containing 2DHG of low density is presented. The figure demonstrates that positively charged excitons consisting of two holes and one electron show in external magnetic field an analogous behaviour to that of their negatively charged counterparts. At zero magnetic field, the PL lines of the exciton (X) and the singlet state of the positively charged trion (X_s^+) are detected (the lower curve in Fig. 2). The binding energy of X_s^+ is 3.1 meV [Ast02a]. The triplet state of the positively charged exciton (X_t^+) is not observed at zero magnetic field. However, in high magnetic fields it becomes bound and with further increase of magnetic fields X_t^+ is detectable even at the lowest energy side of PL spectra (upper curves in Fig. 2).

4. Energy and spin structure of trion states

In order to extract parameters of PL lines of excitons and trions (energy and intensity) with high accuracy a Gaussian fitting of PL spectra has been done. Inhomogeneous broadening of excitonic lines was suggested to dominat at low electron (hole) concentration [Ast01]. Therefore, the linewidths of excitons and trions were assumed to be independent of the magnetic field which noticeably reduces the number of fitting parameters.

Figure 3 shows the energies of the PL maxima in $ZnSe/Zn_{0.82}Be_{0.08}Mg_{0.10}Se$ QW containing 2DEG *vs* magnetic field. The exciton transition shows a regular behaviour with diamagnetic shift split by two well-resolved spin components. From the exciton spin splitting the exciton g factor g_X +0.9 was evaluated. The electron g factor in ZnSe QW's is g_e=+1.1 [Yak00]. It has been measured by spin-flip Raman scattering [Ast02a] and equals the values reported for bulk ZnSe. Using the definition $g_X = g_{hh} - g_e$ [Sir97] we get a value of the heavy-hole g factor $g_{hh} = +2.0$.

We use the determined values of the electron and hole g factors to establish a scheme of the exciton and trion energy levels in magnetic fields in ZnSe QW's as presented in Fig. 4 (see also Ref. [Yak01]). Qualitatively, this is similar to the GaAs QW's case [Shi95a, Van01]. We assume that the g factors are identical for exciton and trion states. The assumption is based on a relatively large splitting between the light-hole and the heavy-hole states in ZnSe-based QW's (e.g. Δ_{hh-lh}=18 meV in the relevant QW) precluding strong variations in mixing of these states by external magnetic fields. This is also confirmed by linear dependence of the exciton spin splitting on magnetic fields presented in Ref. [Ast02a]. Note that the case of ZnSe QW's reported here differs qualitatively from the situation in wide GaAs QW's, where a strong field dependence of the exciton g factor and different spin splitting for excitons and trions have been found [Gla99] due to a different mixing of heavy-hole and light-hole within these states [Riv00].

Based on the scheme the energy of optically inactive excitons can be restored as shown by dotted curves in Figs. 3 and 5. The exchange splitting between optically active and optically inactive excitons does not exceed 0.5 mev and was neglected. The detailed analysis of the exciton states in studied QW's (diamagnetic shift, Zeeman splitting and g factors) in high magnetic fields was reported in Ref. [Ast02a].

According to the scheme, the singlet trion state X_s^- is split with the hole g factor g_{hh}. However, after recombination of the electron-hole pair the electron, which is left in the final state, can have two different (up- and down-) electron spin projections. As a result, the splitting of the optical transitions of the singlet trion state is determined by $g_{hh} - g_e$, which is equal to g_X. In fact, the spin splitting of the singlet state shown in Fig. 3 turns out to be similar to the exciton splitting, except for a deviation from linear field dependence in the field range 25-40 T. The latter could be caused by the crossover of the σ^+ polarized component with the triplet state and is considered in Sec. 6.

The triplet trion state has six spin sublevels (see Fig. 4). According to the spin optical selection rule $\Delta S_z = \pm 1$, four of them (with $S_z = \pm 1/2, \pm 3/2$) are optically active and can decay radiatively. Sometimes such states are called "bright" states. Note that this should not be confused with the appearance of "dark" and "bright" triplet states because of the orbital selection rule [Dzu00]. Two other states with $S_z = \pm 5/2$ could optically decay only by a spin-scattering assisted process. When spins are conserved, these triplet states are optically inactive, no matter what orbital wave functions they have. It follows from the scheme of Fig. 4 that only two PL lines can correspond to the optically active triplet states. The reason is that the energies of transitions 3 and 4 coincide. The same holds true for transitions 5 and 6. For the studied ZnSe QW the expected spin splitting for the optically active triplet in PL is $g_{hh} - g_e = +0.9$, i.e. it is the same as for the exciton and the singlet trion state. Experimentally, however, no spin splitting has been detected for the PL line assigned to the triplet state (Figs. 3 and 5). A separation of the triplet line from the exciton transition increases relatively fast with magnetic field and at about 30 T the triplet crosses the upper spin component of the singlet. Such behaviour is in qualitative agreement with theoretical calculations [Whi97, Woj00, Riv00, Yak01].

Energy dependencies of PL maxima in $ZnSe/Zn_{0.89}Mg_{0.11}S_{0.18}Se_{0.82}$ QW containing 2DHG are presented in Fig. 5. Generally, the behaviour of positively charged excitons in external magnetic field is similar to that of negatively charged excitons (compare with Fig. 3) and therefore does not need to be described it in details. We would like to emphasis here two distinctions. First, the triplet state of positively charged exciton X_t^+ approaches the singlet state X_s^+ at lower magnetic fields (20-30 T) than those for negatively charged excitons (25-35 T). Second, a difference in QW width and barrier material results in a difference of the heavy-hole g factor and, therefore, in a difference of excitonic Zeeman splitting shown in Figs. 3 and 5. An analysis of data on spin-flip Raman scattering and exciton splitting *vs* magnetic field (see Ref. [Ast02a]) gives values for the heavy-hole $g_{hh} = +1.8$ and the exciton $g_X = +0.7$ g factors.

5. Binding energy of trion singlet state

The binding energy of trions (E_B^T) *vs* magnetic field is plotted in Fig. 6. In order to avoid uncertainties caused by spin splittings, data for the center of gravity of exciton and trion spin doublets are given.

Binding energies of X_s^- in all studied QW's show a monotonic increase with magnetic fields and a tendency to saturation in high fields B>25 T. The increase is stronger in wider QW's with smaller E_B^T, it amounts, e.g., to 150% in a 190 Å QW and is only 35% in a 48 Å QW. Being more compact in narrow QW's the singlet state becomes less sensitive to compression by external magnetic fields. Qualitatively $E_B^T(B)$ dependencies for X^- from Fig. 6 are consistent with theoretical predictions for the singlet state [Whi97, Woj00, Riv00, Yak01].

Magnetic field dependence of the singlet state of the positively charged exciton X_s^+ differs drastically from the X_s^- behaviour. The binding energy of X_s^+ shows no dependence on magnetic fields for B<6 T and decreases by 25% at higher fields (see open circles in Fig. 6). In the field range 23-32 T the singlet X_s^+ state line shows irregular behaviour caused by its crossover with the triplet state. These results will be discussed in next section. A qualitatively different behaviour of X_s^- and X_s^+ states in magnetic fields has been established first for GaAs QW's [Gla99]. Our results confirm this for ZnSe-based QW's. We are not aware of any theoretical attempts to model X_s^+ behaviour in magnetic fields. However, it is clear that the difference in the magnetic field behaviour of X_s^- and X_s^+ binding energies is due to a very different structure of wave functions of these complexes (see discussion in Ref. [Ser01]). X^- is constructed of two light particles (electrons) rotated around one heavy particle (hole). This complex has one center and the magnetic field will confine the electrons around the hole, thus increasing the binding energy. In contrast, X^+ has two heavy particles, i.e. two centers, and one light particle moving between them. In this case shrinking of the electron wave function in magnetic fields hinders it from an optimal adjustment between the two centers which results in decreasing binding energy of the X^+ complex. Note that this picture can be applicable at not too high magnetic fields, when the Coulomb energies exceed the hole cyclotron energy.

It is seen from Fig. 6 that the binding energy of the triplet state of the negatively charged exciton grows faster than that of the singlet state. In 67 Å QW (stars) it increases from unbound state in zero magnetic field to 6.8 meV in magnetic field of 45 T. The crossover of singlet and triplet states is expected in magnetic field of about 50 T. Such a behaviour is in good qualitative agreement with theoretical prediction [Yak01]. But from computations the crossover between X_s^- and X_t^- occurs at magnetic fields as high as 250 T. Possible reasons for such discrepancy are discussed in Ref. [Yak01].

The binding energy of triplet state of the positively charged exciton shows similar behaviour to that of negatively charged excitons (crosses in Fig. 6). Being unbound in the absence of magnetic field, binding energy of X_t^+ achieves value of 5.9 meV in magnetic field of 45 T. The crossover between X_s^+ and X_t^+ occurs in magnetic field of 25 T, which is about twice smaller then that happens for X_s^- and X_t^-. Note that the comparison was done for different QW's. In spite of this, we believe that ratio of binding energy of singlet and triplet states should be only slightly dependent on QW width and such distinction in magnetic fields when crossover occurs is mainly due to different structure of wave functions of X^+ and X^-, as was discussed above.

6. Anticrossing of singlet and triplet states

The solid line in Fig. 3 traces an expected energy dependence of the X_s^- PL line for the σ^+ polarized component. It was calculated from the energy dependence of the same line, but detected in the σ^- polarization (solid circles in Fig. 3), and shifted by the exciton Zeeman splitting, which can directly be measured for excitons from the same spectra (open and solid squares). The calculated energy dependence of X_s^- in the σ^+ polarization coincides with the measured one in magnetic fields below 25 T (open circles). In magnetic fields higher than 37 T it is also in good agreement – but with points, labelled as X_t^- (open triangles). Such behaviour looks like an *anticrossing* (AC) of X_s^- and X_t^- states. This is also supported by intensity dependence of the low- (circles) and high (triangles) energy lines of the singlet and triplet states shown in insert of Fig. 3. The exchange of intensities between these lines is also clearly seen, with the intensities being equal in magnetic field of 35 T. We did not find indications to the AC in the σ^- polarization in the studied range of magnetic fields. Because of the Zeeman splitting contribution the AC is expected to be shifted to higher magnetic fields (of about 50-60 T). The repulsion in AC (in magnetic field of 35 T), which gives the strength of interaction between triplet and singlet states, is 1.5 mev, which is about 30% of the X_s^- binding energy of 5.5 meV.

Note, that a negligibly small spin splitting is observed for X_t^- states (Fig. 3). Currently, we do not understand this: basing on a simple spin scheme (Fig. 4) optical transitions for the spin-allowed triplet states should also split like these for the exciton. One possible and rather speculative explanation is that we observe in fact the optically forbidden (i.e., spin-forbidden) transition of triplet sublevel that becomes visible due to interaction with an optically active X_s^- singlet state.

We also checked a possibility if the effect could be explained in terms of interaction of the singlet trion state X_s^- and optically forbidden exciton X. As shown in Fig. 3, optically inactive excitons (dotted lines) are energetically displaced from X_s^- and X_t^- states. Basing on this, we believe that optically forbidden excitons could play only a minor role in explanation of anticrossing behaviour.

Figure 5 shows that the singlet-triplet anticrossing is also observed for positively charged excitons (the repulsion between lines and exchange of intensities are present). Two AC's are clearly detected at 25 and 30 T, for σ^+ and σ^- polarizations. Repulsion in anticrossing is 0.5 meV, i.e. about 20% of the X_s^+ binding energy. It is three times smaller than that for X^-, but binding energies of X_s^- and X_s^+ differ only by a factor two. The observation of singlet-triplet anticrossings both for positively and negatively charged trions in ZnSe-based structures makes us believe that we are dealing with a general phenomenon rather than with an individual property of a selected sample. Very recently we have found the anticrossing behaviour of singlet and triplet states of negatively charged excitons in CdTe-based QW's.

Let us now discuss the physical mechanisms, which can be responsible for interaction (mixing) of the triplet and singlet trion states. We would like to stress that the observation of *anti*crossing is rather unexpected because the singlet and triplet states should have different (electron) permutational symmetries. Therefore, the singlet-triplet interaction means that the electron spin symmetry of charged excitons must be broken by some agent. At least three mechanisms that might be responsible for this can be suggested: (i) The first mechanism has an "intrinsic" origin and might be operational for an *isolated* trion state in the absence of any scattering due to disorder or presence of a 2DEG. This mechanism is related to the *electron-hole exchange* interaction [Steb76] that leads to formation of X^- multiplets characterized by the total angular momentum J

(total spin of electrons S_e=1/2 being combined with that of the hole J_h=3/2). Theoretical consideration of this mechanism that fully take into account the complex valence band structure show, however, that the states that emerge from the (electron) singlet and triplet X^- states belong to different J-multiplets and, therefore, still have different symmetries and cannot anticross. (ii) Second mechanism could be a combination of the e-h exchange interaction in X^- with the scattering (by disorder in QW's and crystal imperfections). The latter breaks the spherical symmetry so that the states from different J-multiplets can be admixed. Our analysis shows, however, that if the disorder scattering is described by a spin-independent potential, the second mechanism does not still lead to the singlet-triplet anticrossing. (iii) The third mechanism that can be indicated is of extrinsic origin and is related to the interaction of X^- with the 2DEG. In this case the exchange of an electron from X^- with electron from the 2DEG, when they have different spin orientations, would switch the trion between the singlet and triplet state and, consequently, introduce mixing (coupling) of these states. One should expect that the coupling efficiency (i.e. repulsion in anticrossing) increases with increasing electron density and strongly depends on the polarization of the 2DEG in magnetic fields. Some indications that the X^--2DEG interaction might play an important role in determining optics of charged trions X^- were found in [Sanv02]. In our opinion, the X^--2DEG interaction can also be responsible for the observed singlet-triplet anticrossing. In order to achieve a better understanding of this interaction and its possible interplay with disorder further theoretical investigations and experimental studies are required.

Acknowledgements. The authors are thankful to R. A. Suris for useful discussions. The work was supported by the Deutsche Forschungsgemeinschaft (Grant Nos. Os98/6 and SFB 410) as well asby the Russian Foundation for Basic Research (Grant No. 00-02-04020). At SUNY Buffalo it was supported by the NSF grant DMR-0203560.

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Figure captions

Fig 1. PL spectra of 67 Å thick $ZnSe/Zn_{0.82}Be_{0.08}Mg_{0.10}Se$ QW, containing 2DEG of low density, measured in zero magnetic field and in magnetic field of 20 T. The spectra are shifted for clarity.

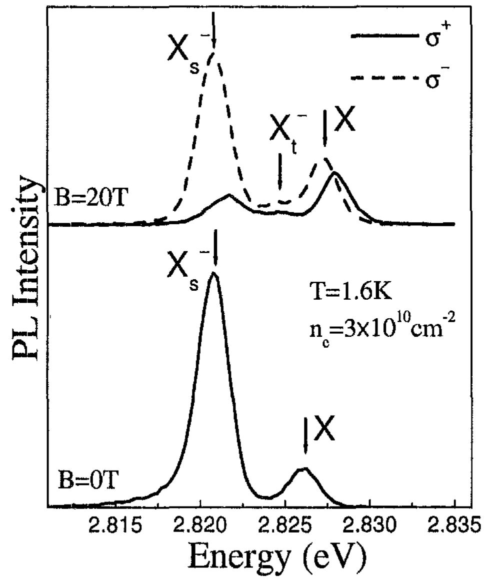
Fig 2. PL spectra of 105 Å thick $ZnSe/Zn_{0.89}Mg_{0.11}S_{0.18}Se_{0.82}$ QW, containing 2DHG of low density, measured in zero magnetic field and in magnetic field of 35 T. The spectra are shifted for clarity.

Fig 3. The energy of neutral exciton X, and energy of singlet and triplet states of negatively charged exciton X_s^- and X_t^- (shown both σ^+ and σ^- circular components) as function of magnetic field in $ZnSe/Zn_{0.82}Be_{0.08}Mg_{0.10}Se$ QW. Dotted lines show the energy of optically inactive states of the neutral exciton, solid line traces an expected behaviour of singlet state of the negatively charged exciton measured in σ^+ polarization. Inset shows an intensity of X_s^- and X_t^- PL lines detected in σ^+ circular component as a function of magnetic field.

Fig 4. Schema for the energy levels of singlet and triplet trion states in ZnSe QW's in external magnetic fields. The inset contains the schema for the exciton. Optically allowed transitions are shown by arrows: solid for σ^+ polarization and dashed for σ^- polarization.

Fig 5. The energy of neutral exciton, and energy of singlet and triplet states of positively charged exciton X_s^+ and X_t^+ (shown both σ^+ and σ^- circular components) as function of magnetic field in $ZnSe/Zn_{0.89}Mg_{0.11}S_{0.18}Se_{0.82}$ QW. Intensities of these lines are coded by the size of symbols. Dotted lines show the energy of optically inactive states of the neutral exciton

Fig 6 Trion binding energy as a function of magnetic fields for QW's of different width. Negatively charged excitons X_s^- were measured in $ZnSe/Zn_{0.82}Be_{0.08}Mg_{0.10}Se$ QW's: 190 Å (diamonds), 67 Å (squares), and 48 Å (triangles). Positively charged exciton X_s^+ is taken for a 105 Å $ZnSe/Zn_{0.89}Mg_{0.11}S_{0.18}Se_{0.82}$ QW's (circles). Stars and crosses represent binding energy of trion triplet states of X_t^- and X_t^+ respectively.



σ+
σ−
X_s^-
X_t^-
X
B=20T
X_s^-
T=1.6K
n_e=3x10^{10}cm^{-2}
X
B=0T
PL Intensity
2.815
2.820
2.825
2.830
2.835
Energy (eV)

Fig.1

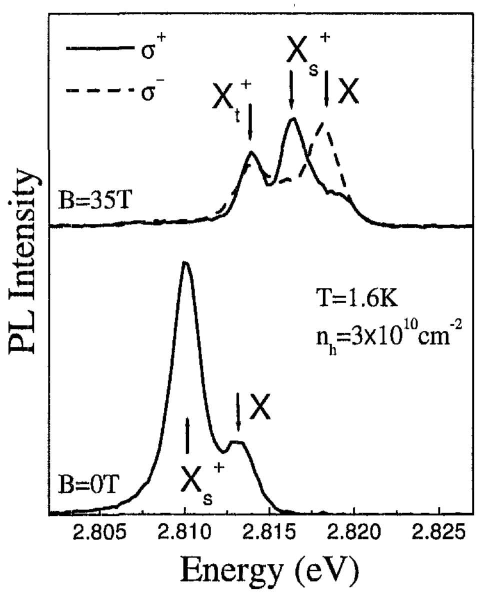
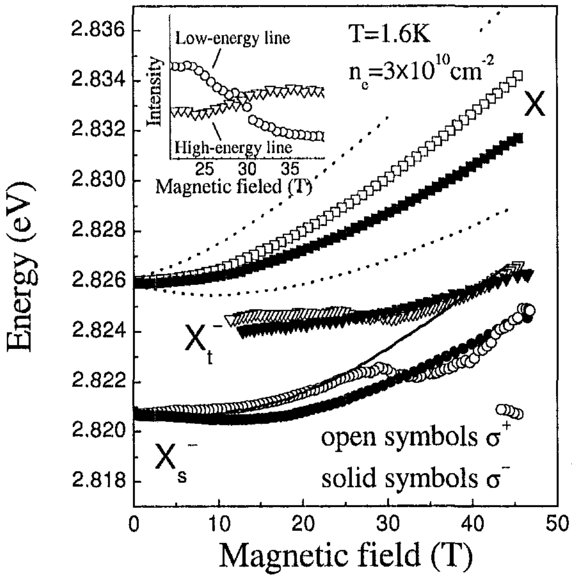

σ+
σ-
X_t^+
X_s^+
X
B=35T
T=1.6K
n_h=3x10^{10}cm^{-2}
X
X_s^+
B=0T
PL Intensity
2.805
2.810
2.815
2.820
2.825
Energy (eV)

Fig.2



T=1.6K
$n_e=3\times10^{10}cm^{-2}$
X
X_t^-
X_s^-
open symbols σ^+
solid symbols σ^-
Low-energy line
High-energy line
Intensity
Magnetic fieled (T)
Energy (eV)
Magnetic field (T)

Fig.3

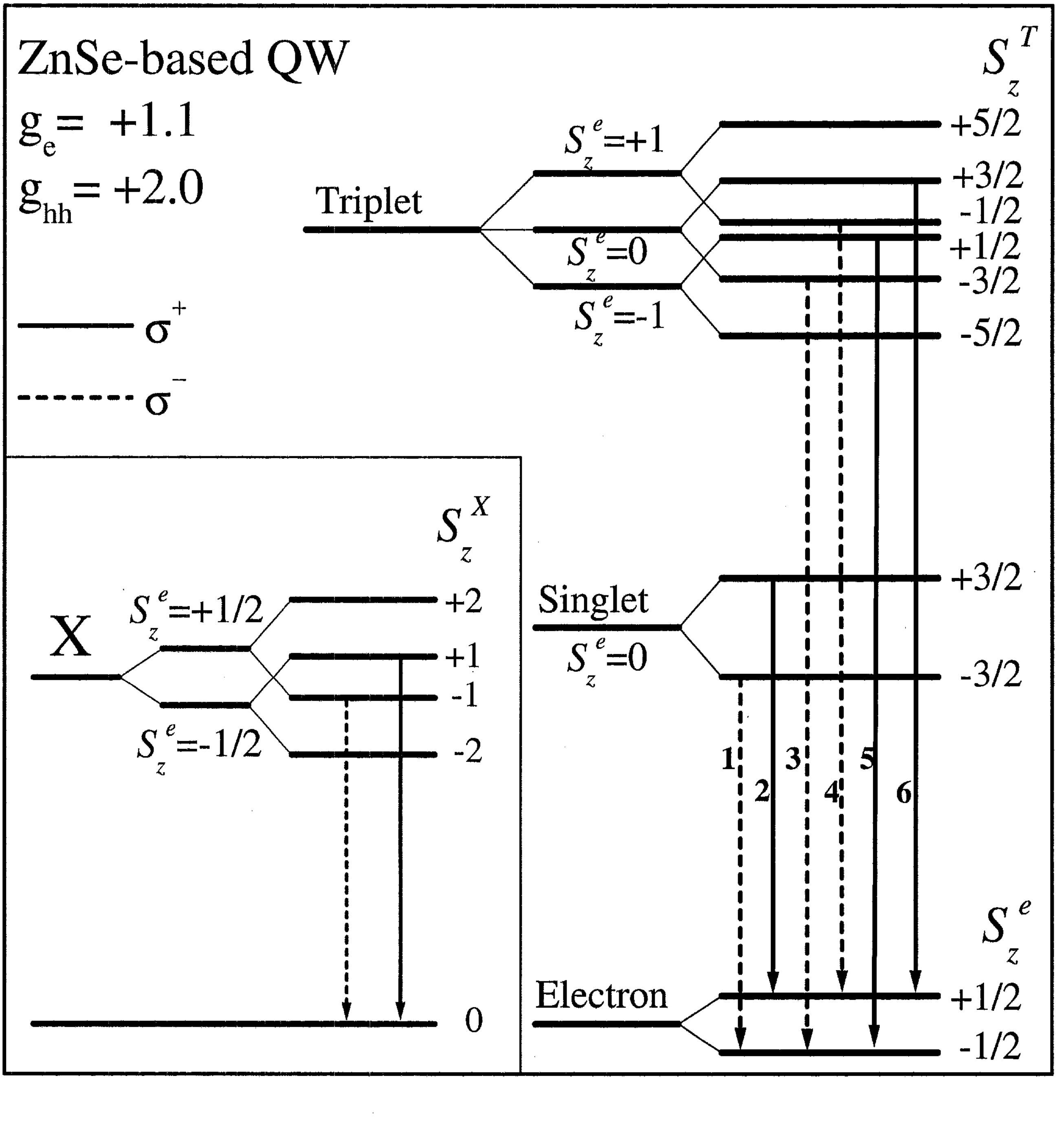
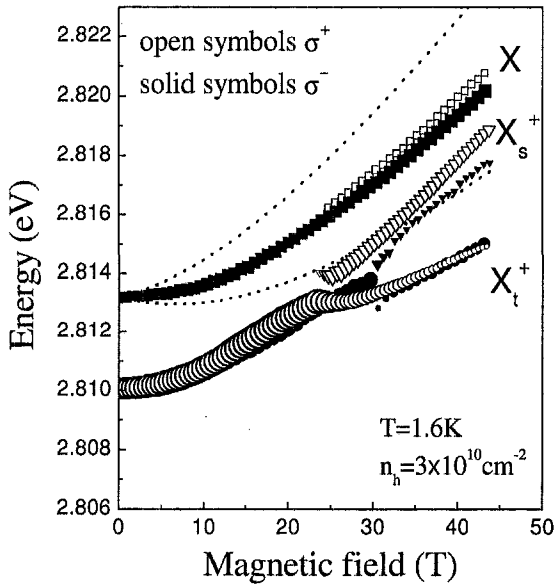
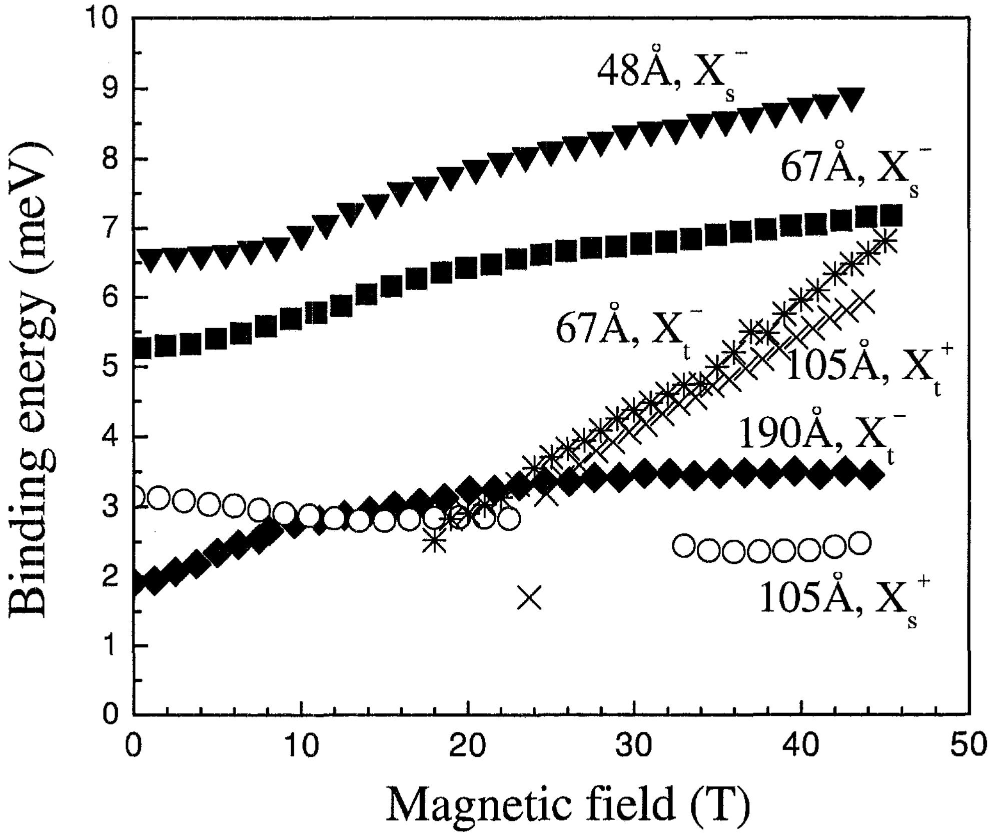

ZnSe-based QW
g_e= +1.1
g_{hh}= +2.0
σ+
σ−
Triplet
S_z^e=+1
S_z^e=0
S_z^e=-1
S_z^T
+5/2
+3/2
-1/2
+1/2
-3/2
-5/2
Singlet
S_z^e=0
+3/2
-3/2
1
2
3
4
5
6
Electron
S_z^e
+1/2
-1/2
X
S_z^X
S_z^e=+1/2
S_z^e=-1/2
+2
+1
-1
-2
0

Fig.4



open symbols σ+
solid symbols σ-
X
X_s+
X_t+
T=1.6K
n_h=3x10^10cm^-2
Energy (eV)
Magnetic field (T)
2.822
2.820
2.818
2.816
2.814
2.812
2.810
2.808
2.806
0
10
20
30
40
50

Fig.5



48Å, X_s^-
67Å, X_s^-
67Å, X_t^-
105Å, X_t^+
190Å, X_t^-
105Å, X_s^+
Binding energy (meV)
Magnetic field (T)
0
10
20
30
40
50
1
2
3
4
5
6
7
8
9

Fig.6